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REMARKS/ARGUMENTS

Claims 1-11, and 13-15 and 21 remain pending in the present application.

Obviousness Rejection of Claims 1-4, 6-11, 13-15 and 21

In the January 8, 2004 Office Action, claims 1-4, 6-11, 13-15 and 21 were rejected under 35 U.S.C. §103(a) for obviousness in view of the combination of Song et al. U.S. Patent No. 5,935,643 or Breault et al. U.S. Patent No. 5,732,463 with Japanese Patent Publication No. 20-138268 or Maricle et al. U.S. Patent No. 4,849,253 and with Koschany et al. U.S. Patent No. 6,183,898 or Japanese Patent Publication No. 9-180729. Applicants submit that none of the foregoing references, either alone or in combination, can render claims 1-4, 6-11, 13-15 and 21 unpatentable for obviousness because non of the references, either alone or in combination, discloses or suggests a continuous method for preparing a ***fluid diffusion layer***, which includes a ***substrate*** and at least one ***loading material*** adhered to the substrate, in which the loading material is ***substantially free of electrocatalyst***.

The remarks in the January 8, 2004 Office Action appear to have confused a fluid diffusion ***layer*** with a fluid diffusion ***electrode***. In a proton exchange membrane (PEM) fuel cell, a membrane electrode assembly (MEA) has an ion exchange membrane interposed between two electrically conductive layers. The electrodes typically include a fluid diffusion layer and an electrocatalyst. In assembling the MEA, however, the catalyst layer can be disposed as a layer either on the ion exchange membrane or on the fluid diffusion layer. If the catalyst layer is disposed on the fluid diffusion layer, then the structure is referred to as a fluid

diffusion ***electrode***. *See* paragraph [0002] of corresponding Patent Application Publication No. 2002/0192383 ("The electrodes typically comprise a fluid diffusion layer and an electrocatalyst.").

The applicants' pending claims, on the other hand, define "a continuous method for preparing a fluid diffusion layer comprising a substrate and at least one loading material", in which the loading material is "substantially free of electrocatalyst". None of the pending claims defines a method of preparing a fluid diffusion ***electrode***, although the fluid diffusion layer prepared using the applicants' claimed method could be employed in the preparation of an electrode.

Moreover, it would be improper to combine the teachings of references that disclose the coating of a catalyst layer to prepare a fluid diffusion ***electrode*** with methods of preparing a fluid diffusion ***layer***. The utility of the catalyst layer would be defeated by the applying to the substrate a loading composition that is "substantially free of electrocatalyst", as recited in the present claims.

The references cited in the Office Action generally describe the general formation of a fluid diffusion layer (sometimes referred to as an electrode support), but in only a perfunctory manner and certainly not according to the applicants' claimed method. For example, the Song patent states, at column 3, lines 9-17, that:

> An electrode catalyst slurry and an electrode support are prepared in separate ways. For the electrode support, carbon paper is waterproofed by immersing it in a waterproofing solution mixed with water, drying in the air, and sintering at a high temperature. .... The preparation of the electrode support is completed by the waterproofing.

Breault is also silent as to the preparation of a fluid diffusion layer, stating in its background section that the fluid diffusion layer is a "carbon paper substrate" (column 1, line 25). The fluid diffusion layer employed in Breault's sole working example is a "Teflon wetproofed carbon paper" (column 5, lines 47-48).

The '268 Japanese publication describes coating a catalyst layer onto an electrolyte (that is, an ion exchange membrane; see, for example, paragraph 15 from a machine translation of the publication into English on the Japanese Patent Office's website; attachment A hereto). The '268 publication contains no teaching or suggestion as to the preparation of a fluid diffusion layer. Moreover, the '268 publication is directed to overcoming a problem seldomly encountered with fluid diffusion layers, namely, the swelling and shrinking of an ion exchange membrane depending upon its moisture content. When this problem arises, drying of the catalyst layer applied to the ion exchange membrane results in a non-uniform coating and possible cracking as the coated membrane shrinks (see paragraph 25 of the attached JPO machine translation).

As with Song discussed above, Maricle describes a method of preparing a fluid diffusion electrode by applying a catalyst/polymer floc to a substrate. At column 4, lines 43-46, Maricle describes suitable substrates as follows:

> The substrate particularly useful herein are metal screens, expanded metal, porous sinters of carbon or metal, metal felt, or mesh, as well as suitable supports utilizing carbon particles or carbon fibers.

The '729 Japanese publication describes the deposition of a catalyst layer into a fluid diffusion layer (*see* abstract: attachment B hereto). At paragraph 24 of

the attached JPO machine translation, a carbon paper substrate undergoes a heat treatment after impregnation with a polytetrafluoroethylene (PTFE). Apart from this terse description, the '729 Japanese publication contains no further description as to suitable fluid diffusion layers or methods of preparing them.

Koschany is the only cited reference that describes in any detail the application of a substantially catalyst-free loading material onto a substrate to make a fluid diffusion layer. Koschany's substrate starting materials include "very light, not necessarily electrically conductive but mechanically stable support materials which comprise fibers, e.g. in the form of nonwovens, papers or woven fabrics" (column 2, lines 37-40). A suspension is then prepared from an electrically conductive material (*see* column 2, lines 62-64), and:

> The abovementioned support materials are thoroughly soaked with the suspension mixture or the mixture is uniformly applied to the support material so that the support material is essentially ***homogeneously impregnated***.

(Column 3, lines 35-38; emphasis added). Koschany's impregnated substrate is then dried and sintered (see column 3, lines 39-48). Although Koschany, unlike the other references cited in the Office Action, actually describes a method of preparing a fluid diffusion layer, Koschany contains no teaching or suggestion of a compacting step, as explicitly recited in the present claims.

Obviousness Rejection of Claim 5

Claim 5 is dependent on claim 1, and specifies that the substrate is pretreated with a hydrophobic polymer before the loading composition is continuously applied

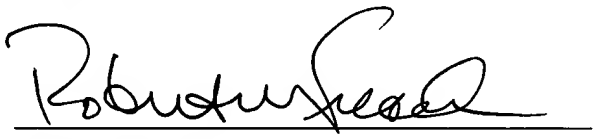
to the substrate. In view of the patentability of claim 1, as established in the preceding remarks, claim 5, which further limits the method of claim 1 to include a substrate pretreatment step, is patentable over the cited references.

* * * * *

In view of the foregoing remarks, applicants submit that claims 1-11, and 13-15 and 21 are allowable. The Examiner is invited to telephone the applicant's undersigned attorney at 312.775.8123 if any unresolved matters remain.

A Petition for One-Month Extension of Time accompanies this paper, as well as a check for the fee for extension within the first month. Please charge any additional fees, and credit any overpayment, incurred in connection with this submission to Deposit Account No. 13-0017.

Respectfully submitted,



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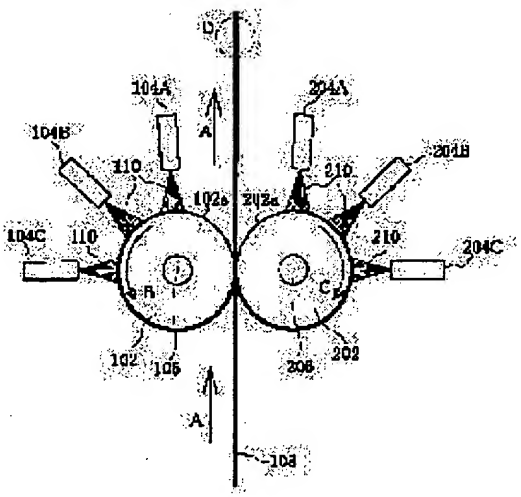
(54) COATING DEVICE AND METHOD USING THE SAME

(57)Abstract:

PROBLEM TO BE SOLVED: To provide a coating device capable of applying a desired material on a continuous thin film through a series of steps without causing a deformation due to coating nonuniformity, clogging, a lack of fine texture and swelling.

SOLUTION: Rollers 102 and 202 synchronously rotates almost at a constant rate in the direction of arrows B and C. Spray guns 101A to 104C, 204A to 204C spray suspensions 110 and 210 towards corresponding outer faces 102 a and 202 a to the rollers 102 and 202. Heating parts 106 and 206 heat the outer faces 102a and 202a from the inner side of the rollers 102 and 202. A continuous belt-like electrolyte film 108 is pressurized with a desired pressure from the both sides of the outer faces 102a and 202a of the rollers 102, 202 while rotating.



LEGAL STATUS

[Date of request for examination]

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rejection]

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[Date of requesting appeal against examiner's decision of rejection]

[Date of extinction of right]

* NOTICES *

1.This document has been translated by computer. So the translation may not reflect the original precisely.
2.**** shows the word which can not be translated.
3.In the drawings, any words are not translated.

DETAILED DESCRIPTION

[Detailed Description of the Invention]
[0001]
[Field of the Invention] In case this invention forms the electrode especially used for the thin film which may be swollen with liquid with a polymer electrolyte fuel cell about the technique for applying a desired ingredient, it is used, and it relates to a suitable technique.
[0002]
[Description of the Prior Art] A fuel cell is equipment which changes into electrical energy directly the chemical energy which a fuel has, and is known as equipment which can expect high energy efficiency.
[0003] The polymer electrolyte fuel cell has composition as shown in drawing 6 among such fuel cells. Drawing 6 is the explanatory view showing the outline configuration of a common polymer electrolyte fuel cell. That is, by the solid-state macromolecule mold cell, the electrochemical reaction shown below advances by supplying the fuel gas containing hydrogen, and the oxidation gas containing oxygen to each of the catalyst electrodes 309a and 309b of the pair formed in both sides of an electrolyte membrane 308.
[0004]
$H_2 \rightarrow 2H^+ + 2e^-$ -- (1)
$2H^+ + 2e^- + (1/2)\,O_2 \rightarrow H_2O$ -- (2)
$H_2 + (1/2)\,O_2 \rightarrow H_2O$ -- (3)
[0005] A formula (1) shows the reaction in hydrogen pole side catalyst electrode 309a, a formula (2) shows the reaction in oxygen pole side catalyst electrode 309b, and a formula (3) shows the reaction performed with the whole fuel cell.
[0006] When the electron generated by such reaction by hydrogen pole side catalyst electrode 309a moves to oxygen pole side catalyst electrode 309b through a load 310, as an arrow head shows to drawing 6 , a current flows from hydrogen pole side catalyst electrode 309a to oxygen pole side catalyst electrode 309b through a load 310, and a fuel cell achieves the function as a cell.
[0007] Now, the above-mentioned catalyst electrode is mainly constituted by the carbon particle of a large number which support the catalyst of noble metals etc. on a front face, and change. Such a catalyst electrode is formed by making it mix in the solvent of a request of the above-mentioned powder of a carbon particle, generating suspension, and applying the suspension to both sides of an electrolyte membrane.
[0008] Now, in the former, there were various methods as shown in drawing 7 as a method which applies a desired ingredient to a thin film.
[0009] Drawing 7 (a) is a method called gravure. By this method, the thin film is sent in the direction of a continuous-line arrow head, pinching the continuous thin film and rotating both rollers with the 1st roller 402 which dipped the lower part of that peripheral face in the liquid containing the above-mentioned ingredient, and the 2nd roller 404 in contact with that roller 402. At this time, the above-mentioned ingredient is applied to the inferior surface of tongue of a thin film by plastering a thin film with the above-mentioned liquid attached to that peripheral face by rotation of the 1st roller 402.

[0010] Drawing 7 (b) is a method called a screen. By this method, the above-mentioned ingredient is applied to the top face of a thin film by placing the screen 406 with which the spreading pattern (pattern aligned with the field which should be applied) was formed on a thin film, and extending the liquid which contains the above-mentioned ingredient by the squeegee 408 on that screen 406.
[0011] Drawing 7 (c) is a method called the Ayr knife. By this method, the thin film is sent in the direction of a continuous-line arrow head, twisting the continuous thin film around the roller 410 which accumulated the liquid containing the above-mentioned ingredient in that upper part, and making it rotate that roller 410. At this time, when a thin film passes through eye the above-mentioned liquid pool, the above-mentioned ingredient is applied to the top face of a thin film. Moreover, by this method, thickness with the ingredient still more uniform than the upper part of the above-mentioned roller applied by spraying Ayr and scratching the excessive liquid which remains in the top face of a thin film is secured.
[0012] Drawing 7 (d) is a method called reverse. By this method, the thin film which continued with ********** the liquid which contains the above-mentioned ingredient with the 1st and 2nd rollers 412,414 with the 2nd roller 414 and 3rd roller 416 is pinched, and the thin film is sent in the direction of a continuous-line arrow head by rotating the 3rd roller 416. At this time, the above-mentioned ingredient is applied to the top face of a thin film by rubbing against a thin film the above-mentioned liquid attached to the peripheral face of the 2nd roller 414 by making the reverse sense rotate the 2nd roller 414 in the 3rd roller 416.
[0013] Drawing 7 (e) is a method called a spray. By this method, while sending the continuous thin film in the direction of a continuous-line arrow head, the above-mentioned ingredient is applied to the top face of a thin film by spraying the liquid containing the ingredient described above from the spray gun 418 arranged above a thin film.
[0014]
[Problem(s) to be Solved by the Invention] As mentioned above, as shown in drawing 7 , there were various methods as a conventional method which applies a desired ingredient to a thin film, but when the catalyst electrode used with a polymer electrolyte fuel cell was formed with such a method, there were the following problems.
[0015] As described above, when forming a catalyst electrode, it is necessary to apply to both sides of an electrolyte membrane the suspension which made the powder of a carbon particle mix in a solvent, using an electrolyte membrane as a thin film.
[0016] However, by the gravure method shown in drawing 7 (a), the roller 402 which dipped the lower part of a peripheral face in suspension is rotated, and since he is trying to apply to the inferior surface of tongue of an electrolyte membrane the suspension attached to the peripheral face, generating of coating nonuniformity is unavoidable. Moreover, in order to pull up suspension toward a top from the bottom by rotation of a roller 402, making it adhere to the peripheral face of a roller 402, irregularity is prepared in the peripheral face of a roller 402, and it is necessary to make it suspension stop at this gravure method mostly by the peripheral face of a roller 402 but, and if it does in this way, while carrying out long duration use, a possibility of starting blinding by the carbon particle by which this irregularity is contained in suspension is. Moreover, for this reason, washing etc. needs to make the peripheral face of a roller 402 periodical, and there is also a problem that a maintenance takes time and effort.
[0017] By the screen method shown in drawing 7 (b), since suspension is extended on the screen 406 with which the spreading pattern was formed, while using it for a long time, a possibility of starting blinding by the carbon particle by which a screen 406 is contained in suspension is like the case of a gravure method. Moreover, for this reason, it is necessary to wash a screen 406 periodically but, and a screen 406 may deteriorate by the penetrant remover then used. Moreover, there is also a problem that a maintenance also takes time and effort.
[0018] the Ayr knife method shown in drawing 7 (c) -- blasting of Ayr -- moreover, by the reverse method shown in drawing 7 (d), it is based on the inverse rotation of a roller 414 -- rubbing -- when it will see microscopically, respectively although there is also no coating nonuniformity and it can apply by almost uniform thickness however if compared with a gravure method, there is a problem that the

delicacy of a spreading side is missing.
[0019] By the spray method shown in drawing 7 (e), since suspension is sprayed with the spray gun 418, the coating nonuniformity, the blinding, and the problem of lack of delicacy which had been generated in the above-mentioned all directions type are not generated. However, there are the following problems common to the above-mentioned all directions type including this spray method.
[0020] That is, since the electrolyte membrane used with a polymer electrolyte fuel cell is a high polymer, when it touches suspension, it has the property in which absorb a solvent and the volume expands (swelling). On the other hand, each above-mentioned all directions type containing a spray method has applied suspension directly to the electrolyte membrane which has such a property. Therefore, the electrolyte membrane applied directly [of suspension] will be swollen by the suspension, and it will deform so that it may lenticulate. And after that, even if it makes it dry, deformation of this electrolyte does not return.
[0021] Then, the following approaches can be considered in order to solve such a problem. That is, the sheet made in polytetrafluoroethylene etc. is prepared first and the above-mentioned suspension is applied by which method of the all directions types described above on the sheet. Next, after making it dry, the sheet is stuck to an electrolyte membrane, heat and a pressure are applied, and hot printing of the carbon particle which adhered on the sheet is carried out on an electrolyte membrane. A catalyst electrode can be formed without making an electrolyte membrane transform by taking such an approach.

[0022] However, since there is nothing if the sheet which is not continuation is not used when taking such an approach, even when applying suspension to the sheet and which method is used, it is necessary to perform baton operation for every sheet. Moreover, it is divided into 2 of the process which applies suspension, and the process which performs hot printing processes, and each process must be performed separately. Therefore, there is a problem that a manufacturing facility becomes complicated and a manufacturing cost becomes high.
[0023] Therefore, without solving the trouble of the above-mentioned conventional technique and producing coating nonuniformity, blinding, lack of delicacy, and deformation by swelling, the purposes of this invention are a series of processes, and are to offer the coater or the method of application which can apply a desired ingredient to the continuous thin film.
[0024]
[The means for solving a technical problem, and its operation and effectiveness] In order to attain a part of above-mentioned purpose [at least], the coater of this invention Being a coater for applying a desired ingredient to the thin film which may be swollen with liquid, and rotating by the peripheral face Let it be a summary to have a mixed liquor adhesion means to make the mixed liquor which mixes the roller which pressurizes said continuous thin film, and said ingredient and desired liquid, and changes adhere to said peripheral face of said roller, and a heating means to heat said peripheral face of said roller to which said mixed liquor adhered.
[0025] Thus, a mixed liquor adhesion means makes the mixed liquor which is mixed with an ingredient and liquid and changes adhere to the peripheral face of a roller in the coater of this invention. A heating means heats the peripheral face of the roller to which mixed liquor adhered. With this heating, among the mixed liquor adhering to the peripheral face of a roller, liquid evaporates in general and an ingredient remains to the peripheral face of a roller. Therefore, if a thin film is pressurized by the peripheral face while a roller rotates, hot printing (hotpress) of the ingredient which remained to the peripheral face of a roller will be carried out on the surface of a thin film by the pressure and heat from a peripheral face.
[0026] Therefore, according to the coater of this invention, it becomes possible to realize all the processes of adhesion, desiccation, an imprint, and a press with the equipment of one unit. Moreover, since not a sheet but the continuous thin film can be treated, it is not necessary to perform baton operation for every sheet like before, and can respond by continuous running. Therefore, a manufacturing facility is easy, ends and can hold down a manufacturing cost at a low price. Moreover, since mixed liquor is not directly applied to a thin film, a thin film swells and does not deform.

[0027] As for said mixed liquor adhesion means, in the coater of this invention, it is desirable to have a blasting means to spray said mixed liquor on said peripheral face of said roller.
[0028] Since mixed liquor will adhere to the peripheral face of a roller by blasting by having such a blasting means, coating nonuniformity does not occur or a spreading side is not fine. Moreover, since it is not necessary to prepare irregularity in the peripheral face of a roller, or to use a screen for it, blinding is not started.
[0029] As for said blasting means, in the coater of above-mentioned this invention which sprayed and was equipped with the means, it is desirable that two or more arrangement is carried out along with the periphery of said roller on the outside of said peripheral face of said roller.
[0030] Thus, two or more laminatings of the above-mentioned ingredient can be carried out on a thin film by arranging two or more blasting means. And it also becomes possible to change the rate of a presentation of an ingredient and the rate of distribution for every layer if needed.
[0031] As for said roller, in the coater of this invention, it is desirable to be arranged at both-sides side of said thin film, respectively.
[0032] Thus, it becomes possible by arranging the roller at both-sides side of a thin film, respectively to apply the above-mentioned ingredient to both sides of a thin film at coincidence.
[0033] As for said roller, in the coater of this invention, it is desirable that coating of said peripheral face is carried out by the matter which has water repellence.
[0034] By coating the peripheral face of a roller in this way, in the case of hot printing, the ingredient which remained to the peripheral face of a roller cannot paste a peripheral face as it is, and an ingredient can be certainly moved on the surface of a thin film.
[0035] As for said heating means, in the coater of this invention, being built in said roller is desirable.
[0036] Thus, magnitude of the whole equipment can be used as a compact by making a heating means build in a roller.
[0037] The process which makes the mixed liquor which the method of application of this invention is the method of application for applying a desired ingredient to the thin film which may be swollen with liquid, and mixes the (a) aforementioned ingredient and desired liquid and changes adhere to the peripheral face of the rotating roller, (b) Let it be a summary to have the process which heats said peripheral face of said roller to which said mixed liquor adhered, and the process which pressurizes said thin film by said peripheral face of said roller by which (c) heating was carried out.
[0038] Therefore, according to the method of application of this invention, the same effectiveness as the same operation as the case of the above-mentioned coater can be done so.
[0039] As for said process (a), in the method of application of this invention, it is desirable to include the process which sprays said mixed liquor on the peripheral face of said roller.
[0040] By having such a blasting process, coating nonuniformity does not occur or a spreading side is not fine. Moreover, since it is not necessary to prepare irregularity in the peripheral face of a roller, or to use a screen for it, blinding is not started.
[0041]
[Embodiment of the Invention] Hereafter, the gestalt of operation of this invention is explained based on an example. Drawing 1 is the sectional view having shown typically the cross section of the principal part in the coater as one example of this invention. The coater of this example is used in order to form a catalyst electrode in both sides of an electrolyte membrane. The catalyst electrode formed in the electrolyte membrane is used as an object for polymer electrolyte fuel cells as shown in drawing 6 .
[0042] The coater shown in drawing 1 is equipped with the spray guns 104A-104C arranged three [at a time], 204A-204C, and the heating unit 106,206 built in the center-of-rotation section of each roller 102,202, respectively along with the periphery of one pair of rollers 102,202, and each roller 102,202, respectively.
[0043] Among these, a roller 102 is for forming the catalyst electrode by the side of the hydrogen pole in a fuel cell, and a roller 202 is for forming the catalyst electrode by the side of an oxygen pole. Coating of the polytetrafluoroethylene is carried out to the peripheral faces 102a and 202a of these rollers 102,202, respectively. Each roller 102,202 is constituted by the roller mechanical component which is

not illustrated, respectively so that synchronous rotation can be mutually carried out at a rate almost fixed to the reverse sense (namely, sense of arrow heads B and C).
[0044] Suspension is supplied to each spray guns 104A-104C, and 204A-204C from the suspension feed zone which is not illustrated, respectively. This suspension makes the powder of the carbon particle which supports the catalyst of noble metals etc. on a front face, and changes mix in a desired solvent (for example, solvent of an alcoholic system), and is generated as mentioned above. In addition, you may make it change the class of catalyst supported by the front face of a carbon particle by the hydrogen pole and oxygen pole side if needed.
[0045] Drawing 2 is the perspective view showing the motion to the roller of the spray gun shown in drawing 1 . It represents with drawing 2 and the motion to the roller 102 of spray gun 104B is expressed with it. In the direction (namely, the direction of an arrow head E) which met the revolving shaft to the corresponding roller 102,202 by the spray gun mechanical component which is not illustrated, as represented and shown in drawing 2 , each spray guns 104A-104C, and 204A-204C are constituted, respectively so that both-way migration can be carried out.
[0046] Each heating unit 106,206 is constituted from the power feed zone which is not illustrated by supply ****** of power, and electric heat, respectively so that the interior of a roller 102,202 to the peripheral faces 102a and 202a can be heated.
[0047] In the above configurations, by this example, as shown in drawing 1 , between peripheral face 102a of a roller 102, and peripheral face 202a of a roller 202, it lets the continuous band-like electrolyte membrane 108 pass, and the electrolyte membrane 108 is pinched by these peripheral faces 102a and 202a.
[0048] Moreover, in the electrolyte membrane feed zone which is not illustrated, the continuous band-like electrolyte membrane 108 is in the condition of having been wound around the coiled form, and is sent out to the sense of an arrow head A towards the above-mentioned roller 102,202 from this electrolyte membrane feed zone. And after minding the above-mentioned roller 102,202, similarly in the electrolyte membrane rolling-up section which is not illustrated, it is rolled round by the coiled form. At this time, an electrolyte membrane 108 is in the condition which fixed tension joined, and is sent by an electrolyte membrane feed zone and the electrolyte membrane rolling-up section at the almost fixed rate doubled with the peripheral speed of a roller 102,202.
[0049] Drawing 3 is a flow chart which shows the rough flow of the spreading processing performed with the coater shown in drawing 1 . In addition, the flow chart shown in drawing 3 does not necessarily show the flow in alignment with time series, and each processing is continuously performed by synchronization. However, since the roller 102,202 is rotating, when its attention is paid to the location of the arbitration on peripheral face 102a of a roller 102,202, and 202a, a part of processings are performed in accordance with time series.
[0050] then, first, as shown in drawing 3 , each roller 102,202 is carrying out synchronous rotation at the rate of about 1 law at the sense of arrow heads B and C as mentioned above (step S102). this time -- an electrolyte membrane 108 -- the peripheral speed of a roller 102,202 -- in all -- it is sent to the sense of an arrow head A at the rate of about 1 law.
[0051] Next, spray guns 104A-104C, and 204A-204C spray the supplied suspension 110,210 toward the peripheral faces 102a and 202a of the corresponding roller 102,202, respectively (step S104). Blasting is turned on / turned off to desired timing, carrying out both-way migration in the direction of an arrow head E (shaft orientations of a roller) by the spray gun mechanical component which is not illustrated, as each spray gun was represented and shown in drawing 2 at this time. Since the roller 102,202 is rotating as it was mentioned above, the blasting pattern 130 of the request by suspension as shown in drawing 2 is drawn on the peripheral faces 102a and 202a of a roller 102,202 by turning on / turning off blasting in this way.
[0052] Moreover, since the sense of rotation of a roller 102,202 is sense of arrow heads B and C as it was shown in drawing 1 , first, suspension 110,210 will be sprayed with spray guns 104A and 204A, next the peripheral faces 102a and 202a of a roller 102,202 will be sprayed with spray guns 104B and 204B, and, finally will be sprayed with spray guns 104C and 204C.

[0053] On the other hand, the heating unit 106,206 is heating the peripheral faces 102a and 202a of a roller 102,202 from the interior of a roller 102,202 (step S108). At this time, the temperature of peripheral faces 102a and 202a heats about 50-150 degrees C of heating units 106,206 so that it may become 100-120 degrees C preferably. By this, the suspension sprayed on the peripheral faces 102a and 202a of a roller 102,202 with the spray gun is dried, the solvent contained in it evaporates and the aggregate of a carbon particle remains on peripheral face 102a and 202a.
[0054] Therefore, if suspension 110,210 is sprayed in order with three spray guns to peripheral faces 102a and 202a as mentioned above Since suspension will newly be sprayed on it and this is repeated, after suspension dries and the aggregate of a carbon particle remains, Finally, on peripheral face 102a of a roller 102,202, and 202a, the laminating of the aggregate of a carbon particle will be carried out over three layers.
[0055] On the other hand, while a roller 102,202 rotates, the band-like electrolyte membrane 108 which are the peripheral faces 102a and 202a, and continues is pressurized by the desired pressure from both sides (step S110). And the peripheral faces 102a and 202a of a roller 102,202 are heated by the heating unit 106,206 as above-mentioned. Therefore, if the part into which the aggregate of the above-mentioned carbon particle on peripheral face 102a and 202a remains by rotation of a roller 102,202 comes to the location which is pinching the electrolyte membrane 108, the aggregate of the carbon particle will be heated and stuck by both sides of an electrolyte membrane 108 by pressure, and hot printing (hotpress) will be carried out by peripheral faces 102a and 202a on the field of an electrolyte membrane 108, respectively. Consequently, a catalyst electrode is formed in both sides of an electrolyte membrane 108 with the aggregate of the carbon particle which carried out the laminating to three layers, respectively.
[0056] Moreover, coating of the polytetrafluoroethylene is carried out as mentioned above in the peripheral faces 102a and 202a of a roller 102,202 at this time. Since polytetrafluoroethylene has water repellence, when it heated and sticks the aggregate of the carbon particle which remained by pressure on an electrolyte membrane 108 by the peripheral faces 102a and 202a, the aggregate of the carbon particle cannot paste peripheral faces 102a and 202a as it is, and it can move the aggregate of a carbon particle on the field of an electrolyte membrane 108 certainly.
[0057] By performing a series of spreading processings continuously as mentioned above, by this example, moreover, hydrogen pole side catalyst electrode 109a and oxygen pole side catalyst electrode 109b can be continuously formed in both sides of an electrolyte membrane 108 at coincidence so that it may expand to drawing 4 and may be shown.
[0058] Drawing 4 is the expanded sectional view having expanded and shown the D section in drawing 1 . In drawing 4 , the catalyst electrodes 109a and 109b with a thickness of about 1-10 micrometers will be formed to the electrolyte membrane 108 with a thickness of about 10-100 micrometers, respectively.
[0059] As explained above, according to this example, it sprays and it becomes possible to realize all the processes of desiccation, an imprint, and a press with the equipment of one unit. Moreover, since the sheet-like not an electrolyte membrane but continuous electrolyte membrane 108 can be treated, it is not necessary to perform baton operation for every sheet like before, and can respond by continuous running. Therefore, a manufacturing facility is easy, ends and can hold down a manufacturing cost at a low price. Moreover, since suspension is not directly applied to an electrolyte membrane 108, an electrolyte membrane 108 swells and does not deform. Moreover, since suspension 110,210 is made to adhere to the peripheral faces 102a and 202a of a roller 102,202 by blasting, coating nonuniformity does not occur or a spreading side is not fine. Since it is not necessary to prepare irregularity in the peripheral faces 102a and 202a of a roller 102,202, or to use a screen for them further again, blinding is not started. Moreover, since the heating unit 106,206 is built in the corresponding roller 102,202, respectively, it can use magnitude of the whole equipment as a compact. Moreover, directly, since the electrolyte membrane 108 is sent in contact with the roller 102,202, it can secure enough a relative location precision of an electrolyte membrane 108 and the catalyst electrodes 109a and 109b formed.
[0060] Now, although he was trying to pinch an electrolyte membrane 108 with the roller 102,202 for forming a catalyst electrode, respectively, you may make it pinch an electrolyte membrane 108 with the

roller for forming the auxiliary roller and catalyst electrode in the above-mentioned example, using an auxiliary roller, as shown in drawing 5 .

[0061] Drawing 5 is the sectional view having shown typically the cross section of the principal part in the modification of the coater of this invention. As shown in drawing 5 , in this modification, it is pinching "Resemble the roller 502 for forming hydrogen pole side catalyst electrode 109a, the auxiliary roller 508, the roller 602 for forming oxygen pole side catalyst electrode 109b by the downstream, while pinching "Be alike", and the auxiliary roller 608" by the upstream [in / for an electrolyte membrane 108 / that feed direction]. And while a roller 502,602 and the auxiliary roller 508,608 rotate to the sense of an arrow head, respectively, the electrolyte membrane 108 is also sent to the sense of an arrow head according to the peripheral speed of a roller 506,606. At this time, a spray gun 504 sprays suspension toward the peripheral face of a roller 502, and a spray gun 604 sprays suspension toward the peripheral face of a roller 602. Moreover, the heating unit 506,606 built in each roller 502,602 is heating the peripheral face of a roller 502,602, respectively. The roller 502 which is pinching the electrolyte membrane 108, the auxiliary roller 508, and a roller 602 and the auxiliary roller 608 are pressurizing the electrolyte membrane 108 by the desired pressure from both sides, respectively, rotating.

[0062] Therefore, also in this modification, it can spray and hydrogen pole side catalyst electrode 109a and oxygen pole side catalyst electrode 109b can be continuously formed in both sides of an electrolyte membrane 108 like [it is possible to perform desiccation, imprint, and a series of spreading processings of a press continuously, and] the example shown in drawing 1 .

[0063] In addition, this invention can be carried out in various modes in the range which is not restricted to the above-mentioned example or the above-mentioned operation gestalt, and does not deviate from the summary.

[0064] Although reference was not made in the above-mentioned example especially about a difference of the suspension supplied to three spray guns For example, by changing the variance of the carbon particle contained in the suspension to supply for every spray gun Since the rate of the presentation of the aggregate of a carbon particle which carried out the laminating to three layers formed in both sides of an electrolyte membrane, and the rate of distribution are changeable for every layer, precision of the catalyst electrode formed can be improved.

[0065] Moreover, especially in the above-mentioned example, although reference was not made about the class of spray gun, as a spray gun, various things, such as air, air loess, and a slit, can be used.

[0066] Moreover, what is necessary is not to limit this invention to this and just to have prepared it at least one in the above-mentioned example, although three spray guns were prepared in one roller.

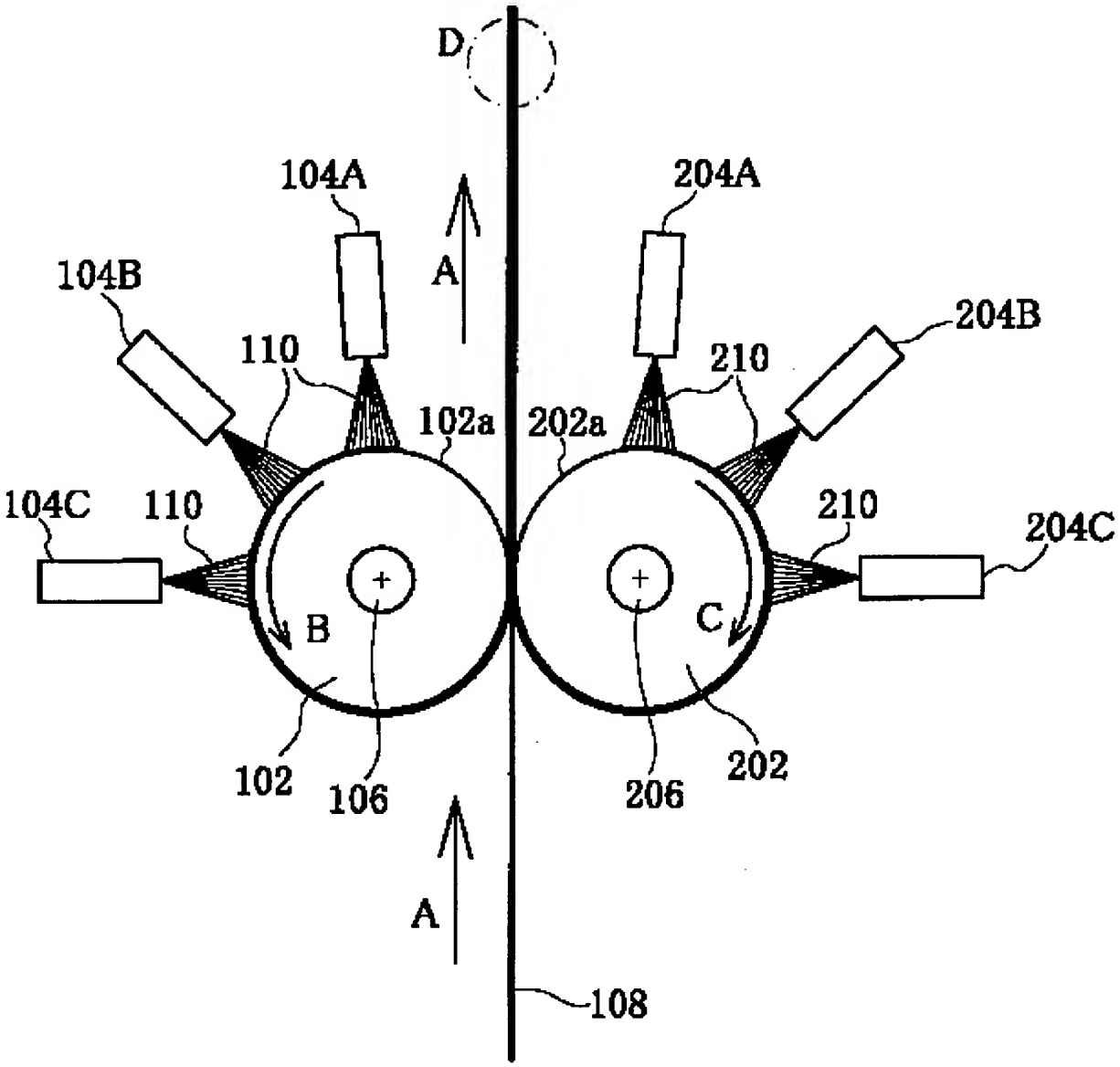
[0067] Moreover, although reference was not made in the above-mentioned example especially about the pressure which a roller 102,202 applies, you may enable it for the pressure regulation section which is not illustrated to adjust the above-mentioned pressure.

[0068] Moreover, this invention is not limited to this, and as long as it is possible to heat the peripheral faces 102a and 202a of a roller 102,202, you may make it prepare it in the exterior of a roller 102,202 in the above-mentioned example, although it was made to build a heating unit 106,206 in a roller 102,202.

[0069] Moreover, although a heating unit 106,206 shall be heated with electric heat, you may make it heat it according to other heat sources in the above-mentioned example.

[0070] Moreover, although carried out in the above-mentioned example as [coat / with polytetrafluoroethylene / the peripheral faces 102a and 202a of a roller 102,202], as long as this invention is matter which is not limited to this and has water repellence, other matter is sufficient as it.

[Translation done.]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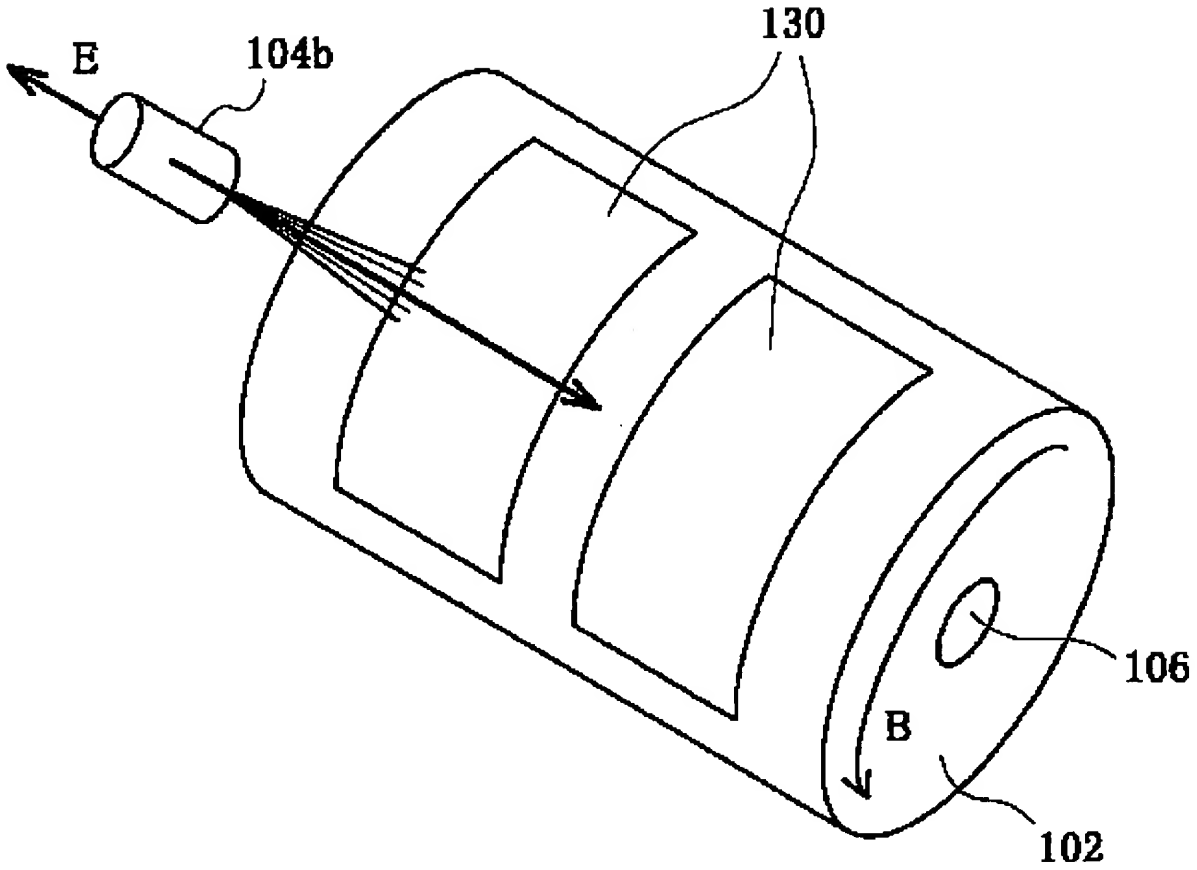
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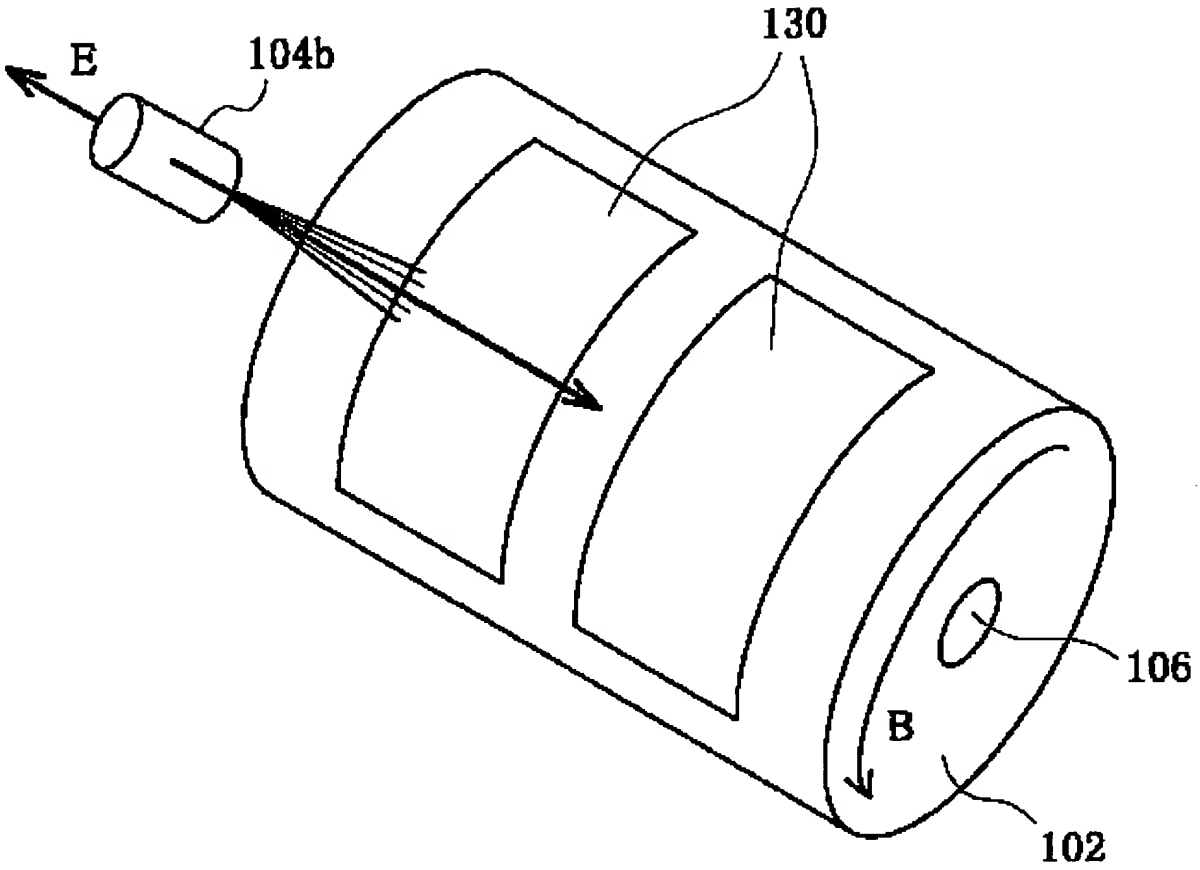
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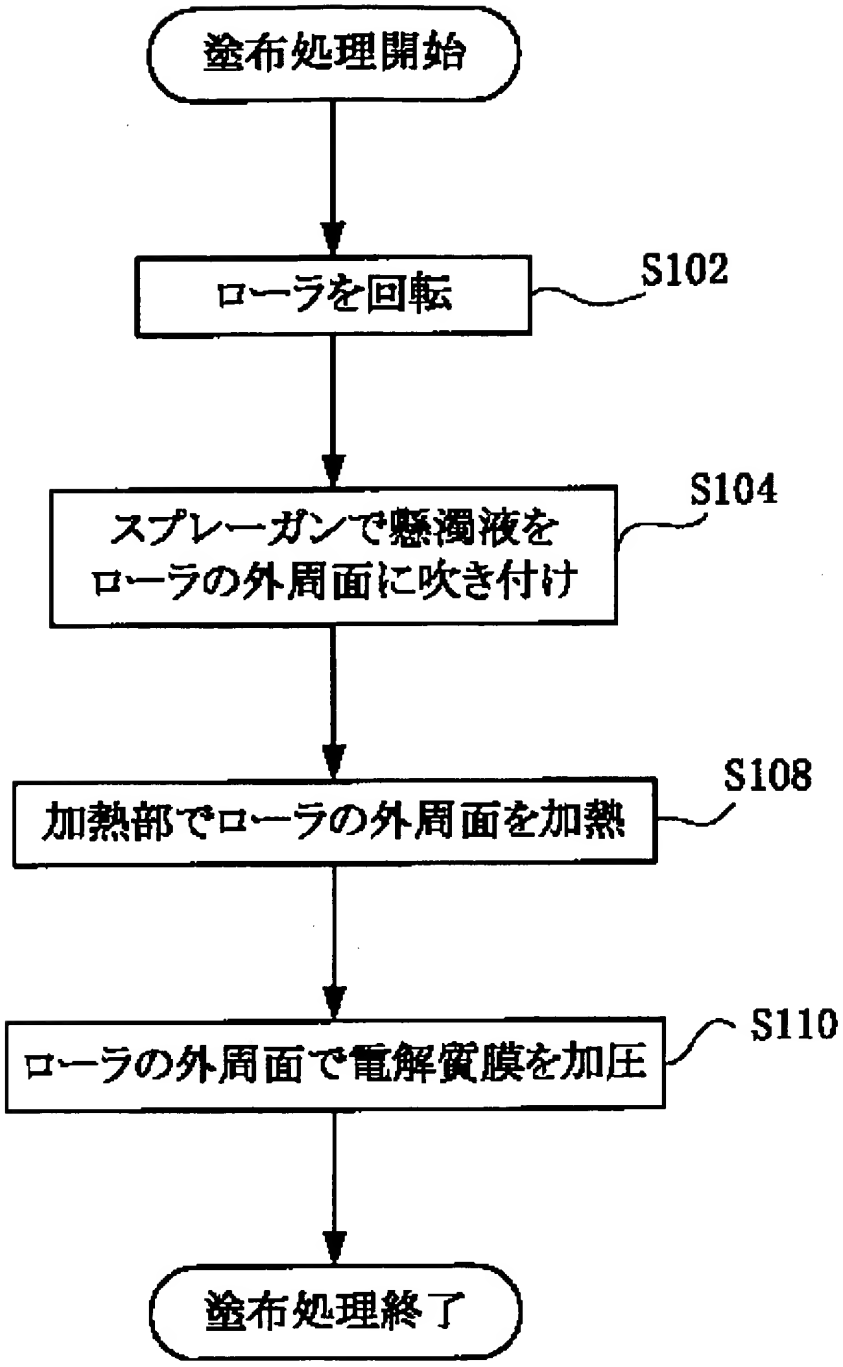
塗布処理開始
ローラを回転
S102
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ローラの外周面に吹き付け
S104
加熱部でローラの外周面を加熱
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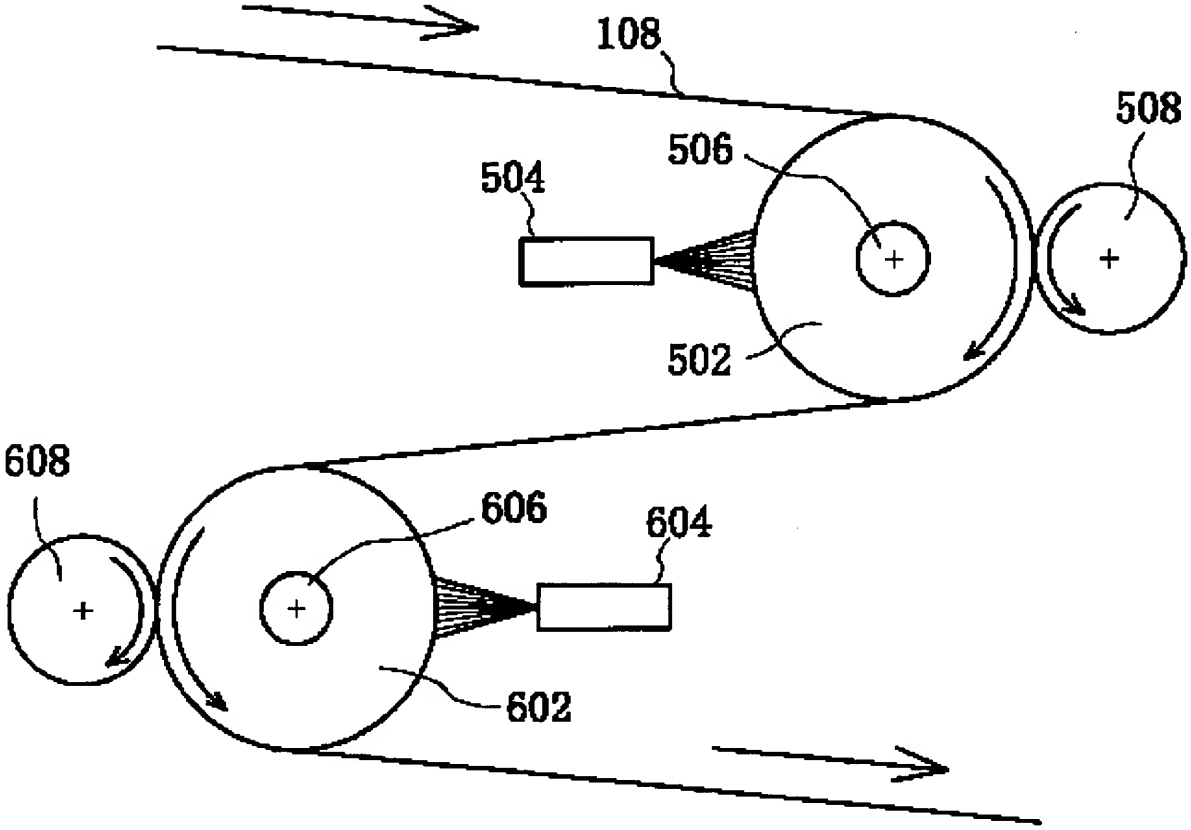
ローラの外周面で電解質膜を加圧
S110
塗布処理終了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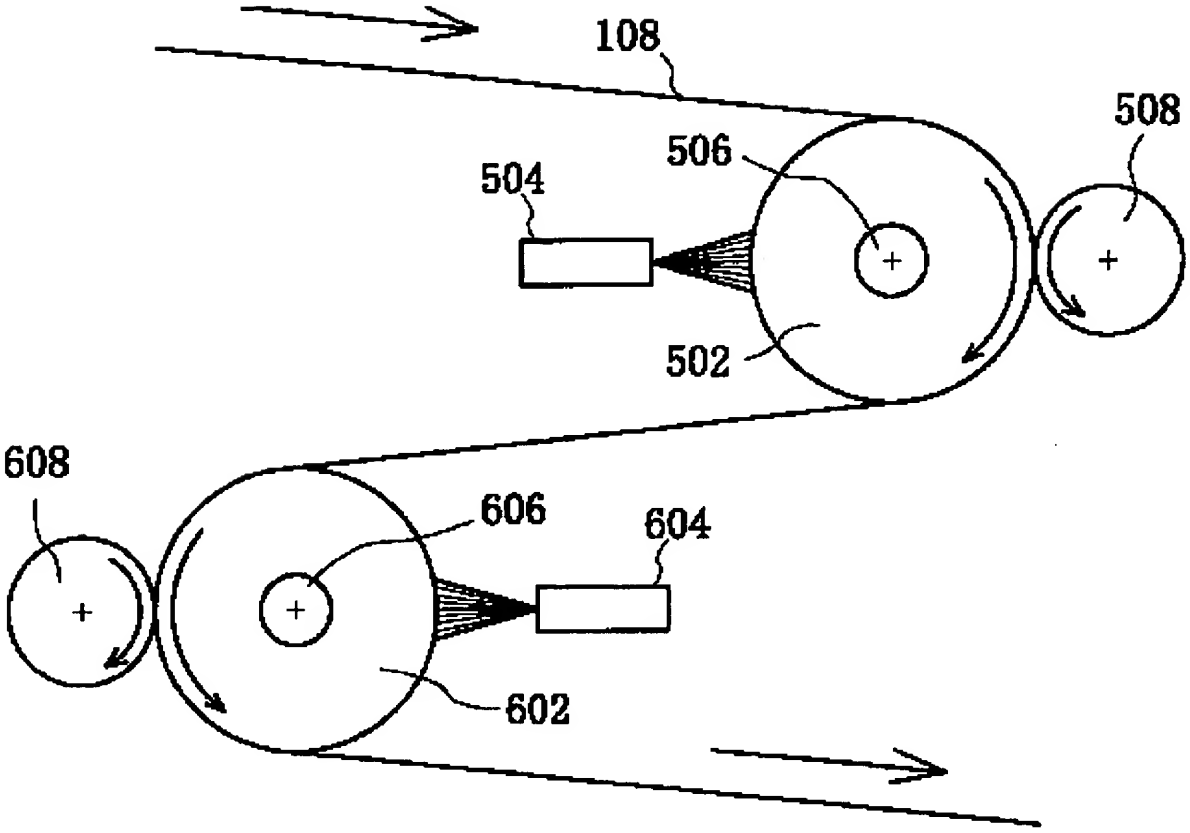
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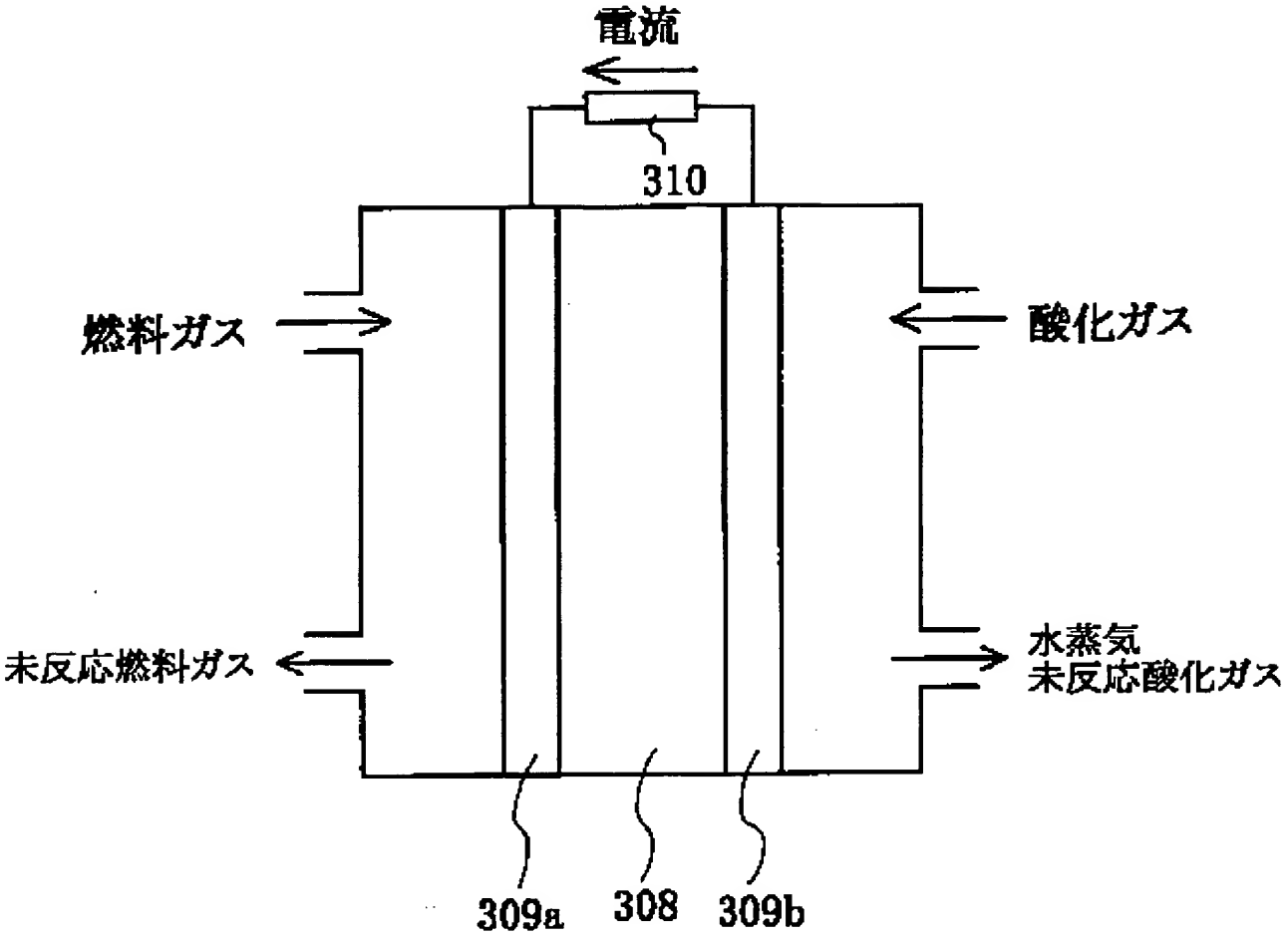
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309a
308
309b

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SEKI TSUTOMU

(54) ELECTRODE FOR SOLID POLYMER FUEL CELL AND MANUFACTURE THEREOF

(57)Abstract:

PROBLEM TO BE SOLVED: To lessen cracking on the surface of a catalyst layer and provide a solid polymer fuel cell with higher performance by removing a solvent by preparatory pressing on completion of deposition of the catalyst layer on a gas diffusion layer by pressure filtration in manufacture processes of an electrode for a solid polymer fuel cell.

SOLUTION: This is an electrode for a solid polymer fuel cell and which is produced by depositing and carrying a catalyst layer containing a catalyst powder and electrolytic substance or a catalyst layer containing a catalyst powder, electrolytic substance, and a water-repelling agent on a gas diffusion layer by pressure filtration. The electrode is produced by depositing a suspension of the catalyst particles on the gas diffusion layer by pressure filtration and then removing an organic solvent by preparatory pressing.

LEGAL STATUS

[Date of request for examination]

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[Patent number]

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[Number of appeal against examiner's decision of rejection]

[Date of requesting appeal against examiner's decision of rejection]

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* NOTICES *

1.This document has been translated by computer. So the translation may not reflect the original precisely.
2.**** shows the word which can not be translated.
3.In the drawings, any words are not translated.

CLAIMS

[Claim(s)]
[Claim 1] The electrode for polymer electrolyte fuel cells which is an electrode for polymer electrolyte fuel cells which makes it deposit and come to support the catalyst bed containing catalyst powder and an electrolyte by pressure filtration on a gaseous diffusion layer, and is characterized by coming to remove a solvent by performing a preliminary press after deposition termination of the catalyst particle suspension by the pressure filtration to a gaseous diffusion layer top.
[Claim 2] The electrode for polymer electrolyte fuel cells which is an electrode for polymer electrolyte fuel cells which makes it deposit and come to support the catalyst bed containing catalyst powder, a water repellent agent, and an electrolyte by pressure filtration on a gaseous diffusion layer, and is characterized by coming to remove a solvent by performing a preliminary press after deposition termination of the catalyst particle suspension by the pressure filtration to a gaseous diffusion layer top.
[Claim 3] The electrode for polymer electrolyte fuel cells according to claim 1 or 2 whose above-mentioned gaseous diffusion layer is carbon paper or ****** carbon paper.
[Claim 4] The electrode for polymer electrolyte fuel cells according to claim 1, 2, or 3 which is the carbon powder with which the above-mentioned catalyst particle supported platinum.
[Claim 5] The electrode for polymer electrolyte fuel cells according to claim 1, 2, 3, or 4 whose above-mentioned electrolyte is resin of a perfluorocarbon-sulfonic-acid system.
[Claim 6] The manufacture approach of the electrode for polymer electrolyte fuel cells characterized by removing a solvent by performing a preliminary press after making the suspension containing a catalyst particle and an electrolyte deposit on up to a gaseous diffusion layer by pressure filtration in manufacturing the electrode for polymer electrolyte fuel cells which makes it deposit and come to support the catalyst bed containing catalyst powder and an electrolyte by pressure filtration on a gaseous diffusion layer.
[Claim 7] The manufacture approach of the electrode for polymer electrolyte fuel cells characterized by removing a solvent by performing a preliminary press after making the suspension containing a catalyst particle, an electrolyte, and a water repellent agent deposit on up to a gaseous diffusion layer by pressure filtration in manufacturing the electrode for polymer electrolyte fuel cells which makes it deposit and come to support the catalyst bed containing catalyst powder, an electrolyte, and a water repellent agent by pressure filtration on a gaseous diffusion layer.
[Claim 8] The manufacture approach of the electrode for polymer electrolyte fuel cells according to claim 6 or 7 that the above-mentioned gaseous diffusion layer is carbon paper or ****** carbon paper.
[Claim 9] The manufacture approach of the electrode for polymer electrolyte fuel cells according to claim 6, 7, or 8 which is the carbon powder with which the above-mentioned catalyst particle supported platinum.
[Claim 10] The manufacture approach of the electrode for polymer electrolyte fuel cells according to claim 6, 7, 8, or 9 that the above-mentioned electrolyte is resin of a perfluorocarbon-sulfonic-acid system.

[Translation done.]

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DETAILED DESCRIPTION

[Detailed Description of the Invention]
[0001]
[Field of the Invention] This invention relates to the electrode for polymer electrolyte fuel cells and its manufacture approach of the format which deposits the catalyst bed containing the catalyst bed which contains (A) catalyst powder and an electrolyte on a gaseous diffusion layer or (B) catalyst powder, an electrolyte, and a water repellent agent, and it makes it come to support in more detail about the electrode for fuel cells, and its manufacture approach.
[0002]
[Description of the Prior Art] A solid-state polyelectrolyte mold fuel cell makes electrochemical reaction cause by having the description at the point which an ion conductor, i.e., an electrolyte, is a solid-state, and is a macromolecule, and the ion-exchange-resin film etc. specifically being used as that solid-state polyelectrolyte, and arranging the two electrodes of a negative electrode and a positive electrode on both sides of this electrolyte membrane, supplying hydrogen to a negative-electrode side, and supplying oxygen or air to a positive-electrode side, and generates the electrical and electric equipment. In this case, as two electrodes of the negative electrode which touches that solid-state polyelectrolyte film, and a positive electrode, in order to promote a reaction in it, there is a thing of the format that the catalyst of platinum, palladium, and others is added and used, and the thing of former versatility has been proposed as a manufacturing method of the electrode of this format.
[0003] For example, the approach of mixing a catalyst particle with ion exchange resin, considering as an electrode sheet, and carrying out thermocompression bonding of this to the ion-exchange-resin film as a solid-state polyelectrolyte is indicated by U.S. Pat. No. 3134697, and the approach of mixing a catalyst particle with polytetrafluoroethylene, considering as an electrode sheet, and carrying out thermocompression bonding of this to the ion-exchange-resin film is indicated by U.S. Pat. No. 3297484 or U.S. Pat. No. 3432355. However, only by joining the solid-state polyelectrolyte film and an electrode sheet as it is by thermocompression bonding etc. in this way, the limit of the reaction site (reaction zone) is carried out to the two-dimensional interface of an electrolyte and an electrode, and there is little substantial active area.
[0004] As one of the technique which improves this, the contact of an electrode material and a solid electrolyte ingredient is made [many], and attaining three-dimensions-ization of a reaction site is proposed. "Electrochemistry" According to 53, No.10 (1985), and P.812-817 NAFION-117 film (the product made from Du Pont --) which is a kind of the perfluorocarbon-sulfonic-acid resin film as solid-state polyelectrolyte film It is manufactured by the process below an outline, while joining a platinum electrode to one side of this NAFION film by the electroless deposition method (penetration method) and considering as a hydrogen pole (an anode, fuel electrode) using a trade name, the oxygen pole (air pole), i.e., the cathode lateral electrode, which constitutes the counter electrode of this electrode.
[0005] First, the carbon powder which supported platinum black powder or 10% of platinum as catalyst powder is used. To this, Amberlite IR-120B(T-3) [styrene divinylbenzene sulfonic acid type resin, Na mold, with a grain size of 30 micrometers powder, product [made from Organo], and trade name], or

NAFION-117[perfluorocarbon-sulfonic-acid resin (H mold), 5% solution in a mixed solvent of fatty alcohol and water, Aldrich Product [made from Chemical] and trade name] is mixed with various mixing ratios.
[0006] Subsequently, after adding and kneading polytetrafluoroethylene by the shape of water suspension to each mixture obtained above, this kneading object is rolled out with roll rolling, and it considers as the shape of a sheet, and they are the temperature of 100 degrees C, and the pressure of 210kg/cm2 to the NAFION film as a solid-state polyelectrolyte after a vacuum drying and about this electrode sheet. A hotpress is carried out. And three-dimensions-ization of an electrode reaction site is attained by mixing ion exchange resin in the oxygen pole joined to the NAFION film as solid-state polyelectrolyte film by one, and the polarization property is raised remarkably there.
[0007] Although each of those electrode sheets is produced by sheet-izing the kneading object of an electrode material by technique, such as rolling, with the above technique, as a method of producing this kind of electrode sheet, the mode which makes this support a catalyst particle is also separately performed using a porous paper or a porous sheet as that base material. For example, this particle is coated with a polyelectrolyte (ion-exchange resin), this particle mixture is sprinkled on the ****** carbon paper as a base material, and it is made to adhere by carrying out a press under heating in JP,4-162365,A using the mixture of the carbon black particle of platinum catalyst support, and a non-supported catalyst carbon black particle as impalpable powder for a sheet-like catalyst bed configuration.
[0008] thus, the layer of the coating catalyst particle to which, as for the obtained electrode sheet, ****** carbon paper formed the gaseous diffusion layer in, and it adhered on this one side -- a catalyst bed -- becoming -- a fuel cell -- it faces incorporating and a catalyst bed side is made to contact a polyelectrolyte film surface As the method of adhesion in up to the gaseous diffusion layer of the catalyst bed, although others, a painting method and the rolling method, a doctor blade method, etc. can apply, in a painting method, large-area-izing is difficult and the rolling method and a doctor blade method not only taking various time and effort but equipment itself is expensive. [method / such / pressing]
[0009] For this reason, as that method of making it adhere to up to gaseous diffusion layers, such as ****** carbon paper of a coating catalyst particle, on the technique (****** carbon paper etc.) adapting especially a filtration format, i.e., those gaseous diffusion layers, this invention person poured out the water solution containing a catalyst particle for catalyst bed formation, paying attention to the technique to filter (suction filtration), developed the result relevant to this previously, and has applied (JP,7-130377,A).
[0010] In the manufacture approach of the electrode for polymer electrolyte fuel cells, invention concerning the above-mentioned application uses ****** carbon paper etc. as a base material, and in applying the suspension which mixed the dispersion of a polytetrafluoroethylene system polymer to the catalyst particle covered with the polyelectrolyte by this in a filtration format, it is characterized by carrying out by distributing the suspension in a dilute sulfuric acid. Although this technique improves this in this way paying attention to that suspension itself, it raises the property of an electrode "pass this process by this", is a total, and has improved the engine performance of a cell sharply.
[0011] By the way, in the above-mentioned filtration process, adhesion of the catalyst particle to the field of ****** carbon paper etc. is made more reliable. Although they it not only merely pours out dispersion liquid such, but can be performed in the so-called suction filtration format decompressed from a lower part, or the format pressurized from the upper part for making the particle mix also inside the field of the ****** carbon paper etc. furthermore etc. For example, a limit is in the scale or magnitude in a NUTTSUE (Buchner funnel) format, and large-area-izing of about [that a uniform layer cannot be formed] and a processing side is difficult.
[0012] Then, this invention person is in charge of manufacturing the electrode for fuel cells which makes it come to support a catalyst bed on a gaseous diffusion layer, in order to solve still such a problem. the solution containing a catalyst particle -- a hollow tube-like object and its top face -- a funnel -- the electrode manufacture approach and equipment of a fuel cell which make a catalyst bed

deposit equally on a gaseous diffusion layer are previously developed by applying pressure filtration using the inferior lamella formed in the ** (Japanese Patent Application No. No. 309931 [six to]). Drawing 1 - drawing 2 show the manufacture approach of this electrode for fuel cells, and one mode of equipment.

[0013] One is a hollow tube-like object among drawing 1 , and a circle configuration like drawing 2 (a) of what did not restrict but carried out the square and the polygon of a pentagon and others is [this cross-section configuration] usable. Although this hollow tube-like object 1 is arranged at a vertical type as it is shown in drawing 2 (a), proper things, such as glass and metal, can be used for it as that quality of the material. As for a superior lamella and 3, an inferior lamella, and 4 and 5 are [two] packing of the upper part and a lower part among drawing 1 , respectively, and 6 is a compressor. Among these, the up-and-down packing 4 and 5 is constituted by the configuration doubled with the configuration of the vertical periphery section of a hollow tube-like object, for example, when a hollow tube-like object is cylindrical, it is constituted in the shape of a circular ring corresponding to the vertical periphery section.

[0014] Moreover, a superior lamella 2 is equipped with the tubing (with a bulb) 7 which introduces the solution to filter, and the tubing (with a bulb) 8 which emits air at the time of excessive pressure, and the tubing 9 which introduces the compressed air from the compressor 6 which raises the internal pressure in a container is connected with it. The solvent exhaust port by which 10 was prepared in the center section of the inferior lamella 3, and 11 are the legs attached in the inferior lamella 3 at one, and 12 is a gaseous diffusion plate which a catalyst bed deposits from a solution. This gaseous diffusion plate 12 will be pinched between the lower opening edge of the hollow cylinder object 1, and packing 5, and will be deposited on that top face by making this into a filter, the solute, i.e., the catalyst particle, in a solution.

[0015] as a dotted line showing an inferior lamella 3 in drawing 2 (b) -- desirable -- a funnel -- it is constituted by the **. Thereby, the solvent after filtration flows smoothly. the top face of an inferior lamella 3 -- such -- a funnel -- in other configurations of hollow tube-like object 1 grade, by constituting in a **, the solvent after ****** and filtration flows smoothly toward a solvent exhaust port. Moreover, even if distribution arises in the thickness of a deposit during the actuation, in a thick part, flow worsens, and since the rate of sedimentation of a catalyst particle falls, it can consider as a layer uniform as a whole. Extent of the inclination can be suitably set up in a required limit, when acquiring such operation effectiveness.

[0016] It faces operating the equipment beyond the outline, the solution containing a catalyst particle is supplied through a conduit 7 from the hold container into the hollow cylinder object 1, the compressed air is introduced by the compressor 6, and the inside of the hollow cylinder object 1 is operated as a pressurization condition. in this case, the funnel of many properties, such as the fluidity of the solution with which extent of that pressurization contains the scales (the path of the hollow barrel 1, height, etc.) of equipment, and a catalyst particle, and reinforcement of gaseous diffusion plate 12 the very thing, and inferior lamella 3 top face, although extent of the ** inclination etc. can be selected more suitably how Usually, in the case of 30cm and about height 5cm, 0.1kg/cm2 of diameters of the hollow cylinder object 1 is carried out below by G (gage pressure), for example.

[0017] According to the electrode manufacture approach and equipment adapting pressure filtration of a more than, it is 2 100cm. Even if it is the above large area, it is uniform and, moreover, the outstanding electrode with the sufficient gaseous diffusion engine performance can be obtained. moreover, still very cheaper compared with other catalyst bed membrane formation equipments according to [there is also an advantage that the pore which the solvent passed serves as a diffusion way of gas, at the time of that manufacture, and] this equipment -- etc. -- the outstanding effectiveness is acquired. Moreover, the base material of the electrode itself is a thing and, as for a gaseous diffusion plate (layer), ****** carbon paper is used preferably as this ingredient. After this ****** carbon paper uses the carbon paper which has predetermined thickness and predetermined porosity and infiltrates the dispersion of for example, a polytetrafluoroethylene system polymer into this, it ****** by carrying out heat treatment.

[0018] Furthermore, the suspension which mixed and obtained ** platinum black particle, the platinum

support carbon black particle, and the solution of a solid-state polyelectrolyte as a solution containing the above-mentioned catalyst particle, ** Although the solution which forms catalyst beds, such as suspension which comes to mix for example, a polytetrafluoroethylene system polymer as a binder (it is also a water repellent agent), is used for the suspension of ** If the thing which made the dilute-sulfuric-acid water solution distribute the suspension like invention of above-mentioned JP,7-130377,A is used, ****** can also acquire both effectiveness for ******.

[0019] By the way, the raw material of the catalyst bed described above is three, catalyst powder, a water repellent agent, and an electrolyte, in these, it considers as a solvent and considers as a slurry, and it is making it deposit on a gaseous diffusion layer by the pressure filtration method, and the effective outstanding effectiveness as mentioned above is acquired [water]. However, it was observed that it is the factor from which a large and small check will occur on the surface of a catalyst bed in process of the evaporation if the catalyst bed obtained in this way although evaporation removal of this was carried out by vacuum evaporation etc. since the water as a solvent was still contained at the deposition termination time by the pressure filtration method is further observed in a detail, and this prevents the cell engine performance.

[0020]

[Problem(s) to be Solved by the Invention] Then, the catalyst bed in which this invention contains (A) catalyst powder and an electrolyte on a gaseous diffusion layer, Or by removing a solvent by performing a preliminary press after the deposition termination in manufacturing the electrode for fuel cells by making the catalyst bed containing (B) catalyst powder, an electrolyte, and a water repellent agent deposit and support by the pressure filtration method The above-mentioned fault is improved, and it improves and aims at offering the electrode for polymer electrolyte fuel cells which has the still higher engine performance, and its manufacture approach.

[0021]

[Means for Solving the Problem] Namely, this invention deposits the catalyst bed containing catalyst powder and an electrolyte by pressure filtration on a gaseous diffusion layer. After deposition termination of the catalyst particle suspension are the electrode for polymer electrolyte fuel cells which it makes it come to support, and according to the pressure filtration to a gaseous diffusion layer top, The electrode for polymer electrolyte fuel cells characterized by coming to remove a solvent by performing a preliminary press is offered. Moreover, the catalyst bed containing catalyst powder, a water repellent agent, and an electrolyte is deposited by pressure filtration on a gaseous diffusion layer. It is the electrode for polymer electrolyte fuel cells which it makes it come to support, and the electrode for polymer electrolyte fuel cells characterized by coming to remove a solvent is offered by performing a preliminary press after deposition termination of the catalyst particle suspension by the pressure filtration to a gaseous diffusion layer top.

[0022] Moreover, this invention offers the manufacture approach of the electrode for polymer electrolyte fuel cells characterized by removing a solvent by performing a preliminary press, after making the suspension containing a catalyst particle and an electrolyte deposit on up to a gaseous diffusion layer by pressure filtration in manufacturing the electrode for polymer electrolyte fuel cells which makes it deposit and come to support the catalyst bed containing catalyst powder and an electrolyte by pressure filtration on a gaseous diffusion layer.

[0023] After making the suspension containing a catalyst particle, an electrolyte, and a water repellent agent deposit on up to a gaseous diffusion layer by pressure filtration in manufacturing the electrode for polymer electrolyte fuel cells which makes it deposit and come furthermore to support the catalyst bed in which this invention contains catalyst powder, an electrolyte, and a water repellent agent on a gaseous diffusion layer by pressure filtration, the manufacture approach of the electrode for polymer electrolyte fuel cells characterized by removing a solvent is offered by performing a preliminary press.

[0024]

[Embodiment of the Invention] the above-mentioned gaseous diffusion layer -- the base material of the electrode itself -- so-called -- it is -- although the porous paper which especially limitation does not have as a gaseous diffusion layer in this invention, and consists of the various quality of the materials, a sheet

(the designation is suitably carried out to the "paper" including this detail in the letter and both), or these can be ******(ed) and used, carbon paper and ****** carbon paper can be used preferably. Among these, ****** carbon paper ****** by carrying out heat treatment, after infiltrating the dispersion of a polytetrafluoroethylene system polymer to this using the carbon paper which has predetermined porosity and thickness. The polymer of a polytetrafluoroethylene system is the semantics containing an others and tetrafluoroethylene hexafluoropropylene copolymer, its other derivatives, etc. here.
[polytetrafluoroethylene / (PTFE)]
[0025] As the above-mentioned catalyst powder, platinum black powder, platinum alloy powder, platinum or the carbon black powder of palladium support, palladium black powder, etc. can be used. Moreover, although there is especially no limitation as the above-mentioned water repellent agent, it is desirable that it is the polymer of a polytetrafluoroethylene system. The polymer of a polytetrafluoroethylene system is the semantics containing an others and tetrafluoroethylene hexafluoropropylene copolymer, its other derivatives, etc. here. [polytetrafluoroethylene] As the above-mentioned electrolyte (polyelectrolyte), it is advantageous to use the resin of the perfluorocarbon-sulfonic-acid resin system which has the engine performance which was excellent as the electrolyte, although various ion exchange resin etc. can be used, and when using the resin film of a perfluorocarbon-sulfonic-acid system as the solid-state polyelectrolyte film especially, it is still more desirable to use the resin of the perfluorocarbon-sulfonic-acid resin system of a same system.
[0026] In this invention, after making it deposit on the porous paper which consists of the various quality of the materials with pressure filtration equipment as shows the aqueous suspension containing the aqueous suspension or (B) catalyst particle, electrolyte, and water repellent agent for the catalyst beds containing these (A) catalyst particle and an electrolyte for catalyst beds preferably to drawing 1 - drawing 2 , a preliminary press removes a solvent, but when the concrete procedure sticks like 1 voice and it states, it is [following] as (a) - (f). (a) For example, remove some solvents (preferably water) by the evaporator after mixing the catalyst particle, water repellent agent, and electrolyte which supported platinum 50% to carbon powder so that a catalyst particle, a water repellent agent, and an electrolyte may be set to 4:3:3 (weight ratio). (b) In this way, mix the obtained mixture by the colloid mill and consider as suspension.
[0027] (c) On the other hand, after infiltrating the dispersion of for example, neo chlorofluocarbon (a tetrafluoroethylene hexafluoropropylene copolymer, the Daikin Industries, LTD. make, trademark) into carbon paper with a% [of porosity] of 75, and a thickness of 0.4mm, perform heat treatment, and prepare so that neo chlorofluocarbon may occupy 20 % of the weight among the amount of whole. It arranges so that the ****** carbon paper obtained by (d) and (c) may be shown as a sign 12 in drawing 1 , and the suspension obtained by above-mentioned (a) - (b) is made to deposit as a catalyst bed by pressure filtration on this ****** carbon paper side. (e) They are after deposition termination and a catalyst bed The temperature of about 100 degrees C, and 50 kgf/cm2 A preliminary press is carried out with extent and a solvent is removed. (f) Press in inter-electrode [which was produced] on both sides of the solid-state polyelectrolyte film, and consider as a cell.
[0028]
[Example] Hereafter, although the example of this invention is explained, of course, this invention is not limited to this example. The equipment of a pressure filtration format as shown in drawing 1 - drawing 2 as equipment used was used. 0.08kg/cm2 of internal pressure at the time of actuation was carried out as G (gage pressure), using a glass hollow tube-like object with a bore [of 15cm], and a height of 8cm as the hollow tube-like object 1.
[0029] (1) After infiltrating the dispersion of neo chlorofluocarbon (a tetrafluoroethylene hexafluoropropylene copolymer, the Daikin Industries, LTD. make, trademark) into carbon paper with a porosity [2 and 75% of porosity] of with a surface area of 180cm, and a thickness of 0.4mm first, heat treatment was performed and the carbon paper which ******(ed) by neo chlorofluocarbon was obtained. In this case, that quantitative rate was prepared so that neo chlorofluocarbon might occupy 20 % of the weight among that amount of whole. (2) On the other hand, the mixed quantitative ratio (weight ratio) of these catalyst particle, polytetrafluoroethylene, and an electrolyte used as the mixture of 4:3:3 the

catalyst particle which supported platinum 50% to the carbon particle, Pori Flon's (polytetrafluoroethylene, the Daikin Industries, LTD. make, trademark) dispersion, and the water solution of the perfluorocarbon-sulfonic-acid resin as an electrolyte. (3) Some solvents(water) were removed by the evaporator about this mixture, the obtained mixture was mixed by the colloid mill, and it considered as suspension.

[0030] (4) subsequently, the ****** carbon paper obtained by (1) is shown as a sign 12 in drawing 1 -- as -- arranging -- setting -- the solution supply in drawing 1 -- the aqueous suspension obtained from the conduit 7 by above-mentioned (2) - (3) -- supplying -- the amount of platinum support -- 1 mg/cm2 It was made to deposit so that it may become. (5) They are after deposition termination and a catalyst bed The temperature of 100 degrees C, and 50 kgf/cm2 The press (preliminary press) was carried out for 10 minutes, the solvent was removed, and the electrode sheet for examples was obtained. When the catalyst bed front face of this electrode was observed with the electron microscope, it turned out that a check is small very few. (6) In this way, make the catalyst bed side of two electrodes contact a polyelectrolyte film surface, insert the perfluorocarbon-sulfonic-acid system resin film of 80 micrometers of thickness into inter-electrode [which was produced / of two sheets], and they are the temperature of 150 degrees C, and pressure 100 kgf/cm2. Under pressurization, after pressing for 60 seconds, this was incorporated and set within the limit for fuel cells, lead wire, a gas pipe, etc. were connected, and it considered as the cell for an example sample offering.

[0031] Example of <<comparison>> The equipment similarly shown in drawing 1 - drawing 2 was used to another side and the ****** carbon paper which made the same with the above (1) and was produced, and the electrode sheet for the examples of a comparison was produced with the application of the pressure filtration method. In this case, it was made the same [with an example] also about a use catalyst particle, a water repellent agent (polytetrafluoroethylene), electrolytic ingredients, and these quantitative rates, and the suspension which was mixed for about 1 hour and obtained this mixture by the colloid mill was used. It replaced with the preliminary press after deposition termination, all were made the same with the case of an example after that except for the point which carried out the vacuum drying at the temperature of 80 degrees C, and it considered as the cell for the example sample offering of a comparison.

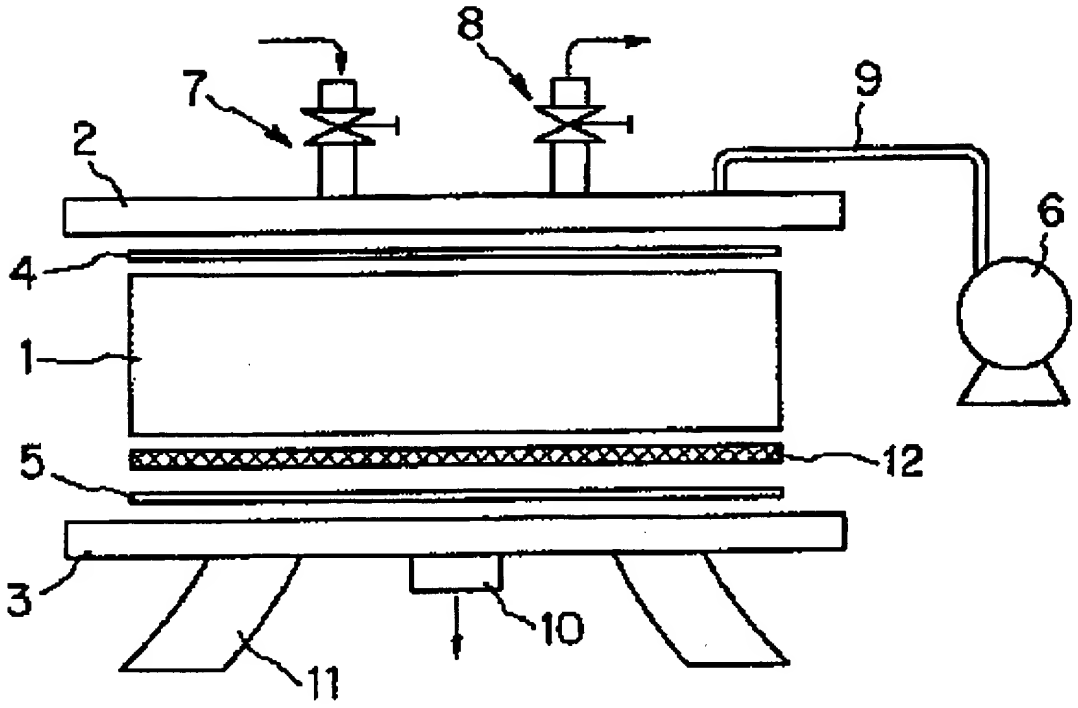
[0032] While using hydrogen as a fuel and supplying this to the anode side using the various sample offering cells manufactured as above, air was supplied to the cathode side. By setting both the supply pressures of this ring main to 2atm(s), hydrogen is 95 degrees C and it humidified at 80 degrees C about air, and the temperature of a cell was kept at 80 degrees C, was operated, and was measured. Drawing 3 shows the relation of the current density and the cel electrical potential difference which were measured about each above sample offering cell.

[0033] Although the cel electrical potential difference shows the **** engine performance fairly only by falling gradually to the increment in current density also by the example sample offering cell of a comparison as drawing 3 , in an example sample offering cell, the fall inclination is further still slower, and it turns out that the cell property is improved further. Thus, according to this invention, it is distinct to improve the property of a polymer electrolyte fuel cell further much more effectively.

[0034]

[Effect of the Invention] According to this invention the above passage, after deposition ending to the gaseous diffusion layer of the catalyst suspension by pressure filtration, by removing the solvent of a catalyst bed with a preliminary press, the check on the front face of a catalyst bed can be lessened, and it can be made small. The condensation of water and the reduction of the contact surface of the electrode by the check and an electrolyte membrane in a check can be prevented by this, and the polymer electrolyte fuel cell which has the further still higher engine performance can be obtained.

[Translation done.]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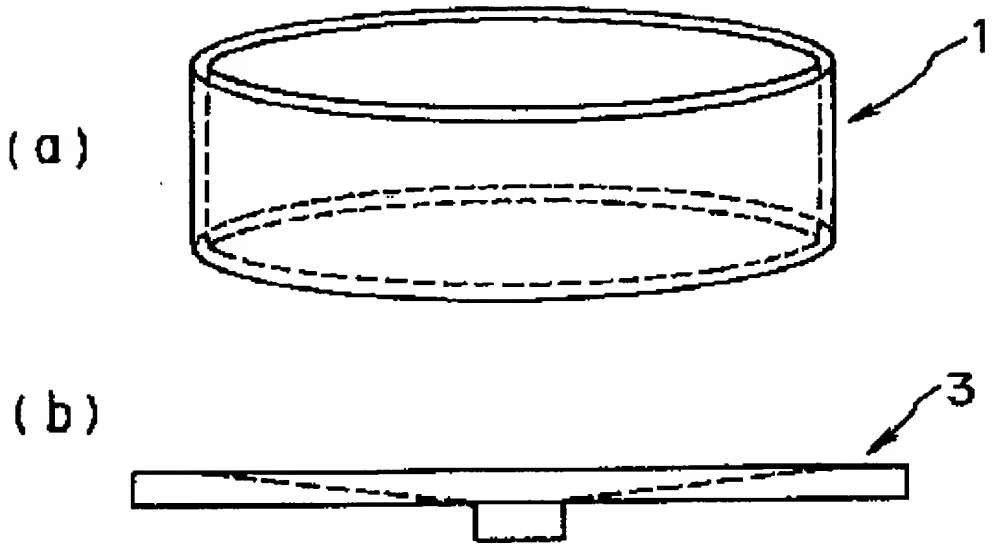
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(b)
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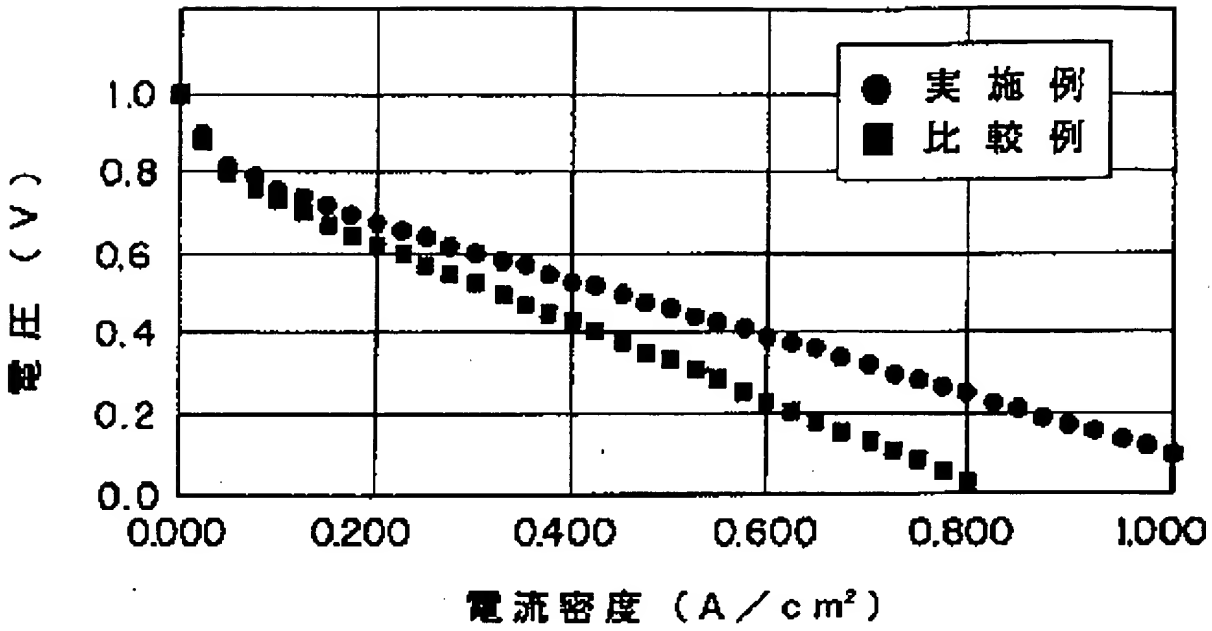
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1.000
電圧（V）
電流密度（A／cm²）